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This report deals with the following areas: (i) screening non-linearities and (ii) coupled intersubband-cyclotron modes in quantum-well structures, (iii) the pressure dependence of two-magnon scattering in antiferromagnetic insulators and (iv) sequential resonant tunneling in superlattices. The studies are primarily experimental focusing on the technique of Raman scattering.

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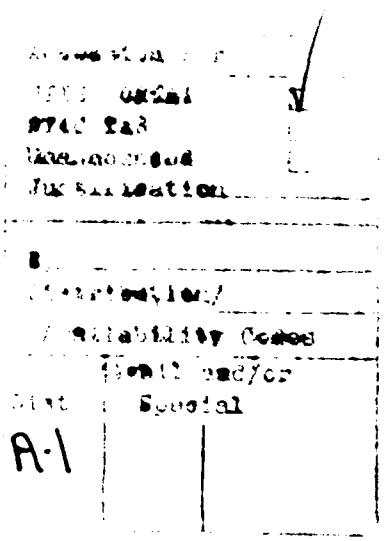

FINAL REPORT
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In this report, we present a brief account of research findings for projects supported by the above ARO Contract [Amount: $230,410.00 (3 yrs)]. Other than the PI, the work involved two graduate as well as two undergraduate students who were partially or fully supported by the grant. Some of the projects benefited from the participation of a visiting Fulbright Scholar.

The main topics of research sponsored by ARO can be divided into four areas dealing with (i) screening nonlinearities and (ii) coupled intersubband-cyclotron modes in quantum-well structures (QWS), (ii) the pressure dependence of two-magnon scattering in antiferromagnetic insulators, and (iv) sequential resonant tunneling in superlattices. Two smaller projects on GaAs/Al/GaAs heterostructures (Publication B2) and on the pressure behavior of optical phonons in In(As,Sb) alloys were also completed. To this, we should add our recent discovery of a giant electro-pleochroism in GaAs quantum-well structures (Publication A8).

A. SCREENING NONLINEARITIES IN QUANTUM-WELL STRUCTURES

This project focused on the screening properties of an electron-hole system confined to a slab; it was motivated by studies of the quantum-confined Stark effect in photoexcited QWS.[1] Results [Publication A2] were obtained by solving the linear-response problem within the Hartree approximation (the consequences, however, are of a more general nature). Our findings reveal a hitherto unrecognized behavior at low densities where the screening charge develops *damped* oscillations. This effect, reflecting remnants of quantum properties in the classical limit, differs significantly from the well-known Friedel oscillations. Extending the treatment beyond linear response, we further studied the capacitive properties of photoexcited quantum wells under various conditions [Publication A3]. Significantly, we found that the strong nonlinearities of the screening coupled with tunneling or absorption nonlinearities can easily lead to negative differential *capacitance* and associated bistability. The calculations shown in Fig. 1 correspond to a situation where the photoexcitation energy is slightly below that of the lowest exciton in the structure;

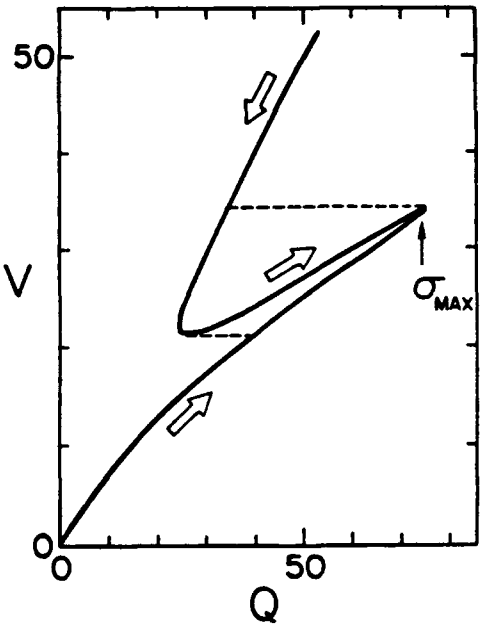

Fig. 1 - V vs Q for near-band-gap photoexcitation. V is the voltage drop across the quantum well in units of $e/2\lambda$ and Q is the external charge in units of $\lambda A/4\pi eL$; L and A are the width and the area of the quantum-well and λ is the energy of the lowest state of the well. σ_{MAX} denotes the largest density, arrows indicate the direction of increasing σ, and dashed lines show the V range of bistability.

the observed behavior reflects the interrelated dependence of the exciton energy and the photoexcited charge-density on the external field. These results - and analogies with materials displaying negative differential resistance (NDR) - led us to the conjecture that photoexcited *multiple* QWS may exhibit *charge-density domains* (as opposed to profiles where the electron-hole density is uniform). Such domains can be viewed as the counterparts of the high and low electric-field domains characteristic of NDR systems.[2] Experimental evidence of charge-density domains in GaAs/(Al,Ga)As structures was reported in Publication B1 It relies on Raman scattering experiments revealing a splitting of an intersubband peak in a narrow range of bias voltages.

B. COUPLED INTERSUBBAND-LANDAU-LEVEL EXCITATIONS

Quasi-two-dimensional electron systems in a magnetic field generally exhibit coupling between the (originally independent) motions perpendicular and parallel to the confinement plane.[3-10] The tilt angle θ, defined by the direction of the field and the normal to the plane, determines the degree of admixture vanishing at $\theta = 0$. In the case of free electrons, tilting results in subband-Landau-level hybridization and associated excitations of mixed character. Experimentally, tilted-field-induced coupling with allowed excitations makes possible for infrared[5-7] and Raman spectroscopy[8] to probe nominally forbidden intersubband or cyclotron resonances. Tilted magnetic fields have also been applied to related studies of confined shallow impurities[8,9] and, more recently, they have helped elucidate the role of reversed spins in the fractional quantum Hall effect.[10]

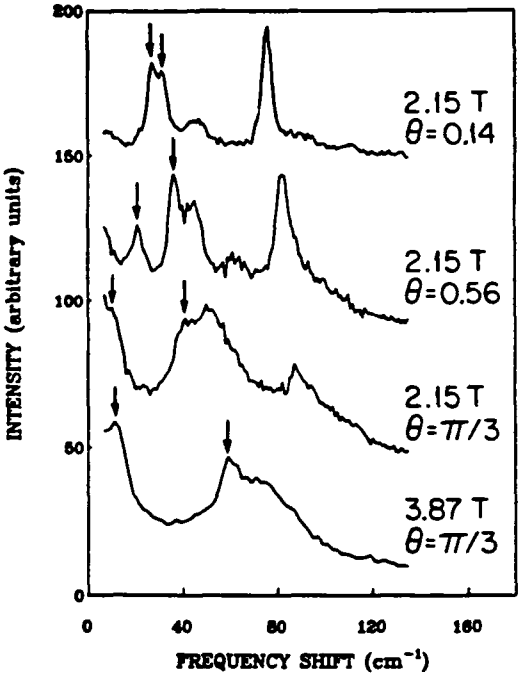

Fig. 2 - Electronic Raman spectra of a quantum well at various angles (θ) and magnetic fields. Arrows denote the combined intersubband - cyclotron resonances. The well-width is 700Å.

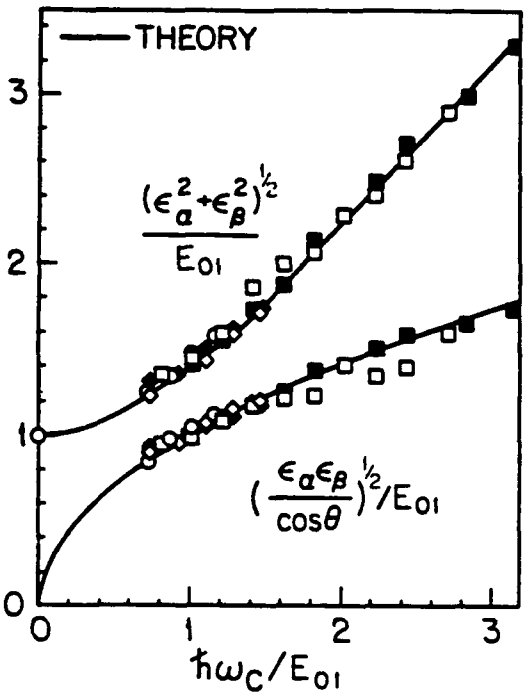

Fig. 3 - Scaled plot of experimental data for three samples (squares, 700Å; diamonds, 460Å; circles, 380Å) at two different angles. Solid (open) symbols are for θ=0.14 ($\theta=\pi/3$). E_{01} is the intersubband energy associated with the lowest states of the well at zero field. The energies of the coupled modes and the cyclotron frequency are, respectively, $\varepsilon_{\alpha,\beta}$ and ω_c.

Following our early work,[8] the ARO project focused on an investigation of intersubband-Landau level coupling in GaAs-QWS involving Raman scattering experiments and various theoretical methods [Publications A6 and B7]. Our experimental approach, using extremely low carrier densities (<10^9cm^{-2} - generated by weak photoexcitation) and QWS, eliminated most of the difficulties plaguing the analysis of infrared studies on space-charge layers.[5,6] The interpretation of the data was further helped by an advantage Raman scattering has over infrared spectroscopy, namely, the ability to discriminate between spin- and charge-density excitations.[11] Typical electronic Raman spectra are reproduced in Fig. 2. Data on various transitions at small tilt angles show very good agreement with results of perturbation theory[3] and, at large angles, with calculations using a finite-basis approximation. In addition, the expected[4] gaps in the excitation spectrum and signatures of the related persistent mixing of levels were clearly observed. As shown by our work, the latter closely relate to the problem of phonon-photon polaritons. Finally, we found that the energies of the lowest-lying coupled modes scale in accordance with predictions of

the parabolic-well model.[4] A scaled plot for three samples and two tilt angles is shown in Fig. 3. Such a surprising behavior relies on the observation that the lowest two branches contain nearly exclusively a combination of the two uncoupled states added to the fact that these states in parabolic and square wells are very similar.

C. PRESSURE-DEPENDENCE OF TWO-MAGNON SCATTERING IN ANTIFERROMAGNETS

Using a diamond-anvil cell, we performed Raman scattering experiments to determine the P (pressure)-dependence of magnon-pair excitations in NiO [Publication A5], α-Fe_2O_3 (hematite) [Publication A1] and $FeBO_3$. All these compounds become antiferromagnetically ordered at low temperatures and exhibit strong two-magnon scattering at ambient pressure (notice that the mechanism for two-magnon excitations in *antiferromagnets* does not rely on spin-orbit coupling unlike that of the much weaker one-magnon scattering).[12] The materials also share in common the fact that their low-lying magnetic states are well accounted for by the Heisenberg hamiltonian (S = 1 in NiO and S = 5/2 for the Fe-compounds) with coupling due to superexchange. Our work was motivated in part by current theories of high-T_C oxide superconductors relying, like Anderson's superexchange theory, on the interplay of charge and spin fluctuations.[13] In this context, our goal was to provide data on archetype antiferromagnetic compounds to contrast with results on the insulating copper oxides. Attempts to correlate the P-dependence of T_C with that of the superexchange constant J have been recently described in the literature.[14] We also note that the dependence of J on P and, thus, on lattice parameters also bears on the possible occurrence of an isotope effect arising from electronic mechanisms.[15] More directly, our experiments were planned to test Bloch's empirical law $J \propto V^{-10/3}$ applying to the V (volume)-dependence of superexchange.[16] It is important to emphasize that the reason why Bloch's relationship works has not been definitively settled and, furthermore, that J-P data are - in general - very scarce. As somehow expected, our Raman measurements show that magnetic excitations react stronger than their vibrational counterparts to pressure changes. Typically, Grüneisen parameters for magnons are a factor of $\approx$ 3 larger than for phonons.

Spectra of NiO at two pressures are shown in Fig. 4. Our measurements, in the range 0-30 GPa, and other results from the literature give $J \propto a^{-\varepsilon}$ with ε = 9.9±0.5 (a is the lattice parameter). This value is consistent with the estimate for $\partial \ln J/\partial P$

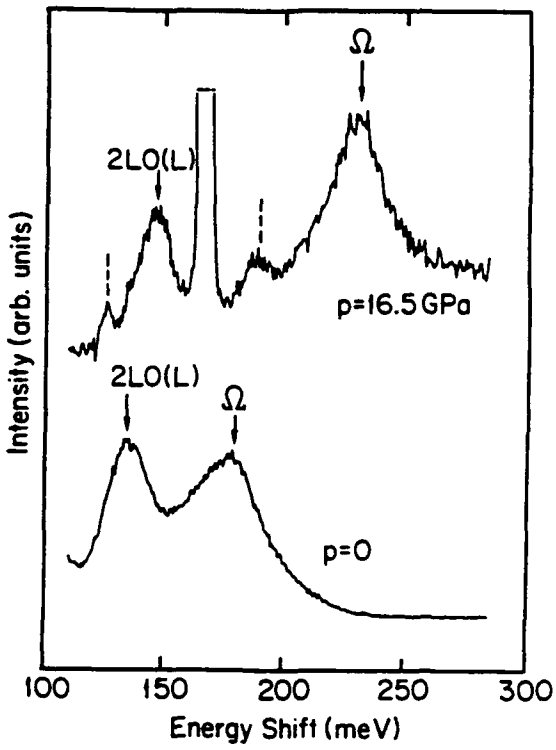

Fig. 4 - Raman spectra of NiO at ambient pressure (bottom trace) and at P=16.5 GPa (top trace). Bands labeled Ω and 2LO(L) are due, respectively, to two-magnon and two-phonon scattering. Dashed lines denote features associated with the pressure transmitting medium. The truncated peak at ~170 meV is the optical phonon of diamond.

in La_2CuO_4 given in the isotope-effect work[15] and with Bloch 10/3 law[16] which is the main assumption in the work on T_C-J correlation.[14] However, given the incomplete identification of the mechanism underlying Bloch's findings, it is not at all obvious why the highly anisotropic and S = 1/2 copper oxides should follow the same trend shown by NiO.

Our experiments on α-Fe_2O_3, combining isotopic substitution with pressure measurements, led to two major results. First, we unequivocally confirmed recent work by McCarty[17] suggesting that the dominant feature in the Raman spectra (at 1320 cm^{-1}) is due to a phonon overtone as opposed to two-magnon scattering. The latter assignment, made in hematite's original Raman work,[18] had resisted the scrutiny of researchers in various fields for over 15 years. Isotopic substitution allowed us to identify the two-magnon peak as a weak and broad feature at ~1525 cm^{-1}, showing a temperature dependence consistent with its magnetic origin.

The study on $FeBO_3$ differs from the previous ones in that it was aimed at probing not $\partial J/\partial a$, but $\partial J/\partial Q$ (Q is a phonon coordinate). To do this, the two-magnon feature was tuned with pressure to resonate with an optical mode of the same symmetry (see Fig. 5). The larger linewidth of the phonon in the crossover region is due to

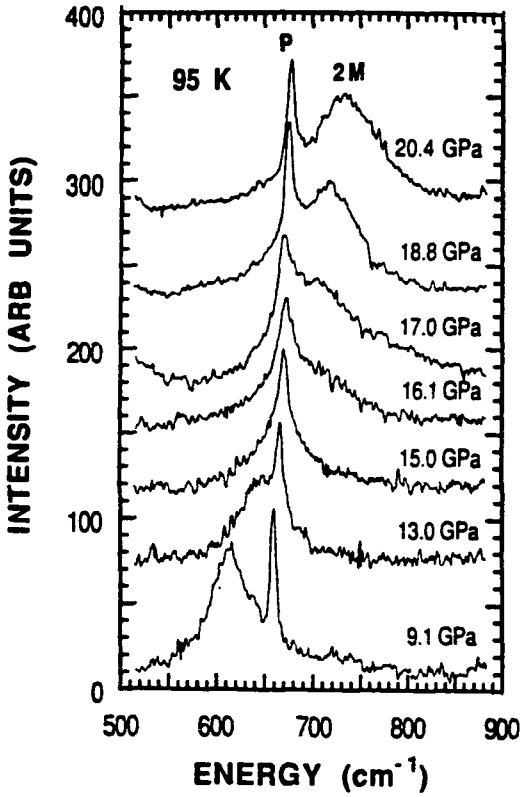

Fig. 5 - Raman spectra of $FeBO_3$ as a function of pressure. The two- magnon feature is labeled 2M. P denotes the E_g phonon.

$\partial J/\partial Q$-induced coupling between the phonon and the continuum of magnon-pairs. Work is now in progress to understand the spectral-lineshape and to obtain quantitative information on the coupling constant.

D. ELECTRIC FIELD DOMAINS IN QUANTUM-WELL STRUCTURES

Here, we describe preliminary studies of sequential resonant tunneling (SRT)[19-27] involving dc-transport, time-resolved PL and Raman scattering measurements. The structure used in the experiments is a nominally *undoped* 131-Å-GaAs/79-Å-$Al_{0.35}Ga_{0.65}As$ superlattice (N = 100 periods) sandwiched between doped GaAs layers in a *p-i-n* configuration. Carriers were generated by means of photoexcitation. A previous PL investigation of the same sample was reported in Ref. 28. Publications B5 and B6 give an account of the Raman data.

The I (photocurrent) - V response of our sample exhibits a strong dependence on photoexcitation intensity.[28] At low power-densities, I shows two maxima at applied voltages $V \cong$ -5V, -12V due to SRT associated with first-second and first-third alignment of electron subbands (the data exhibit no evidence of hole-SRT). Considering the built-in voltage in the structure $V_0 \approx$ 1.5V and the number of periods N, this assignment is consistent with calculations predicting three electron well-states at E_1= 21.5 meV, E_2= 84.75 meV and E_2= 184.13 meV. With increasing power-

density, the response shows a gradual change until the behavior crosses over into that of the domain regime. *I-V* data at high intensities are reproduced in Fig. 6. In the range $5V \leq -V \leq 12V$, the domains with $F = E_{12}/ed$ and $F = E_{13}/ed$ coexist leading to voltage oscillations associated with the motion of the domain boundary. There is also a strong hysteresis in the response, the magnitude of which increases with increasing laser power.

The above results illustrate a significant aspect of SRT in *photoexcited* QWS, namely, the fact that one can continuously monitor the transition from uniform-field to the domain regime. Another important point is the observation of hysteretic behavior. Hysteresis has often been observed in conventional-NDR materials,[2] but it has not been previously reported for QWS.

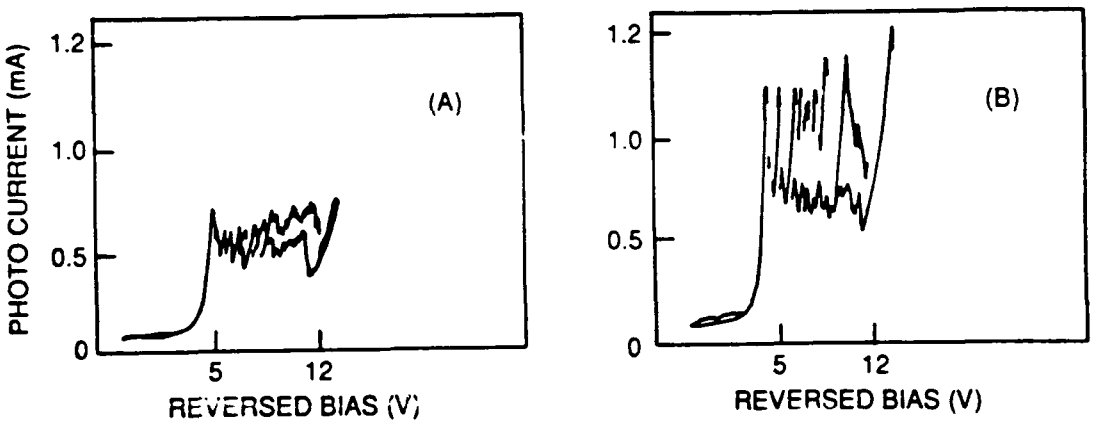

Fig. 6 - Photocurrent (*I*) as a function of applied voltage (*V*) in a GaAs/(Al,Ga)As QWS. The parameters of the structure are described in the text. The laser power is 30 mW for (a) and 130 mW for (b). λ = 5145 Å.

Because of the strong dependence of exciton energies on electric field, particularly for the lowest heavy-hole exciton (HH1), PL is a very useful tool to probe the electric-field profile in QWS. This so-called quantum confined Stark effect[31] has been applied to SRT studies in Refs. 26 and 28. The signature of the presence of domains is the appearance of a doublet in the PL-spectra. Consistent with the above discussion, our sample exhibits two HH1-lines for $5V \leq -V \leq 12V$ such that the peak at higher (lower) energies is due to the low (high) field domain.[28] Moreover, the *V*-dependence of their relative intensity varies according to the expected behavior for the ratio of domain sizes.[28] Here, we notice that two PL-lines should also be observed in the range $-V \leq 5V$ where the coexisting domains are

associated with miniband-conduction and first-second level alignment. However, the calculated separation between the lines is smaller than their widths and, therefore, they cannot be resolved in the spectra.

Our preliminary PL results on domain kinetics are shown in Fig. 7. To understand how domain boundaries form, we applied a step-like voltage profile (i.e., $V = 0$ for $t \leq 0$ and $V = V_0$ at $t > 0$) and measured time-resolved PL-spectra. The results in the figure are for V_0= 7V. The trace at $t = 0$ exhibits a single HH1-peak - corresponding to $F = 0$ throughout the structure - while the data at t = 63 ns (bottom trace) represent the steady-state with two domains where the field equals $\approx E_{12}/ed$ or $\approx E_{13}/ed$. The experiments indicate that the field-profile is nearly uniform for times less than ≈15 ns and that, by then, the whole structure is in the low-field domain. The spectra at later times reflect the motion of the domain boundary associated with the growth of the high-field region. Presently, we do not have a complete understanding of the transient behavior. Nevertheless, it is interesting to point out that the times involved are significantly smaller than well-to-well transit times as measured in QWS of similar parameters.[32]

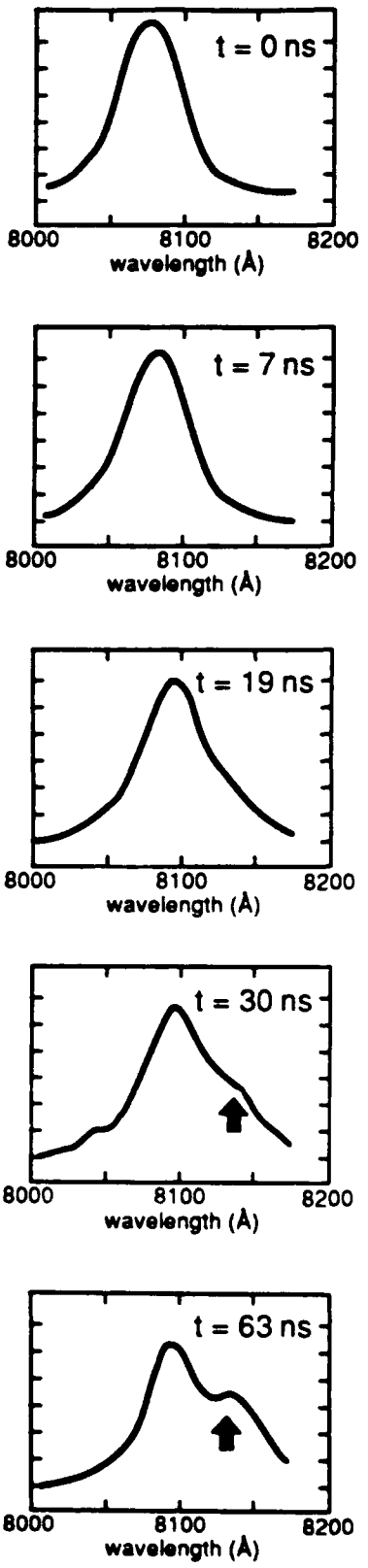

Fig. 7 - Time-resolved PL-spectra of the QWS described in the text. Arrows indicate the HH1-peak associated with the high-field domain.

Our Raman work focused on electron intersubband excitations. Because the energies of these transitions vary with electric field,[1] the spectra give a measure of the QWS field-profile much like that obtained from PL studies. Another significant piece of information provided by Raman - although not by PL - data bears on the electron density in the wells. The latter can be obtained either from the energy separation between spin- and charge-density spectra[11] or from the Raman cross section, being directly proportional to the number of carriers. Here, we notice that, while fields strongly modify the lowest-subband electron-hole overlap and, thus, the PL-intensity, the relevant matrix elements for Raman (involving states at much higher energies) are only weakly affected by electric fields.

The Raman spectra of our sample reveal a feature at $\approx 500\ cm^{-1}$ (62.5 meV) due to electronic transitions from the first to the second subband. This assignment is in agreement with the calculated values for E_1 and E_2 quoted earlier and the estimate for E_{12} from transport data.[28] The behavior of this peak with V is consistent with

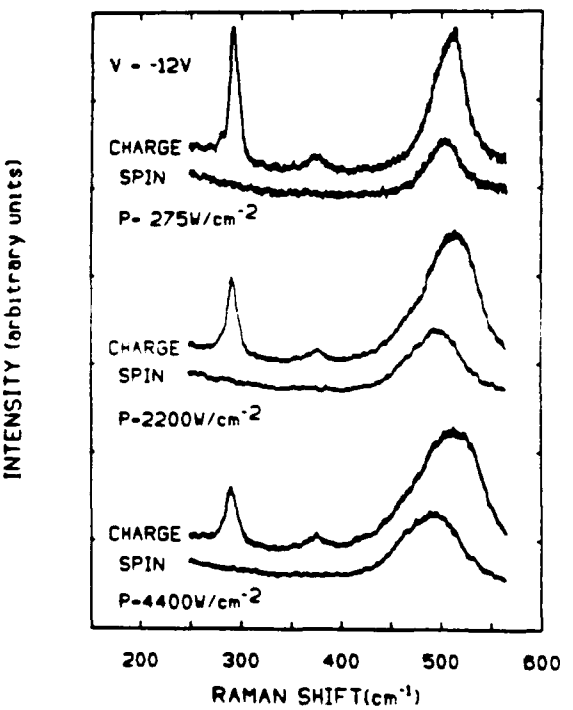

Fig. 8 - Raman spectra at V=-12V as a function of power density P. The top and bottom traces correspond in each case to the charge- and spin-density geometries. The intersubband excitation is the feature at ≈ 500 cm-1. The narrower peak at ≈ 300 cm-1 is the GaAs LO-phonon. For the given value of the external voltage, there is a single domain in the sample. The data show effects due to screening of the external field by the photoexcited carriers.

the PL-results. Unfortunately, the peak is too broad for resolving the expected doublet in the domain regime. The measured dependence of the Raman intensity on V strongly correlates with the photocurrent. This suggests that voltage changes modify mainly the carrier density (as opposed to the transit time) and, moreover, that the domain boundary contains a deficiency of holes. These possibilities are being further

explored at this time.

The PL-work indicates that screening effects are relatively minor for our sample, i.e., the fields are such that $F \gg \sigma$ where σ is the areal carrier density. Albeit weak, these effects manifest themselves in the Raman data. In Fig. 8, we reproduce spectra as a function of power density at a voltage V = -12V bringing the superlattice completely into first-third subband alignment. With increasing excitation intensity, the spin-density scattering, probing essentially the *bare* intersubband energy,[11] exhibits a red shift and an increase in its width. The charge-density line also broadens, but it remains roughly at the same position. Recalling that larger fields lead to larger intersubband transition energies (and that the separation between the two peaks is a measure of the electron density),[11] it is clear that screening by photoexcited carriers is responsible for the observed behavior. That the charge-density excitation hardly moves with P suggests that its alignment is the one that matters for resonant tunneling.

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PUBLICATIONS
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A. JOURNALS

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B. CONFERENCE PROCEEDINGS

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